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Unvented Kerosene Heater Emissions in Mobile Homes Studies on Indoor Air Particles, Semivolatile Organics, Carbon Monoxide, and Mutagenicity

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16. ABSTRACT

This study was conducted to assess human exposure to air pollutants resulting from the use of kerosene heaters in mobile homes. It has been estimated that 15-17 million unvented kerosene heaters have been sold in the United States, and 33% of these heaters have been sold to mobile home residents. The emissions from kerosene heaters can result in high pollutants levels in mobile homes that have a small air volume and low ventilation rate. Eight totally electric mobile homes with no smokers living in the homes were monitored for indoor air particles <10 μm (PM_{10}), semivolatile organics, carbon monoxide (CO), and mutagenicity of semivolatile and particle-phase organics in *Salmonella typhimurium* TA98 without S9 using a microsuspension reverse-mutation assay. Each home was monitored for an average of 6.5 h/day, 3 days/week, for 4 weeks (2 weeks with the heater on and 2 weeks with the heater off) during the heating season of 1989. Indoor air exchange rate, temperature, and humidity were measured. Chemical analyses, including polycyclic aromatic hydrocarbon (PAH) and nitro PAH, also were performed on the indoor air samples from a selected home with the kerosene heater on and off. Increases in CO and organic concentrations resulting from the use of kerosene heaters were found in most homes monitored. High concentrations of both total chromatographable organics (bp between 100-400°C) and gravimetric organics (bp>300°C) were found in homes when the kerosene heaters were used. Overall PM_{10} data from eight homes revealed no significanc kerosene heater effect on PM_{10} concencrations. Two homes, however, showed a significant increase in PM_{10} due to kerosene heater use with concentrations exceeding 100 $\mu g/m^3$ when the heaters were on. Gas chromatography/mass spectrometry analysis data from one home indicated an increase in the concentrations of PAH and nitro PAH due to kerosene heater use. Chemical analysis data also suggested the presence of evaporated, unburned kerosene fuel present in semivolatile organics collected in the XAD samples. When kerosene heaters were on, 56% of the sampling days (in all homes) showed dose-response mutagenic activity and 19% showed mutagenic activity on the heater-off days. In comparison with the U.S. national ambient air standards, four out of the eight heaters investigated in this study emitted pollutants that exceeded the ambient air standards some days. Two homes (Homes 3 and 8) exceeded the 24-h PM_{10} standard and three homes (Homes 1, 4, and 8) exceeded the 1-h peak and/or 8-h average for CO standards. These data suggested that emissions from unvented kerosene heaters can significantly impact indoor air quality in mobile homes and that these emissions contain carcinogenic compounds and can be potentially carcinogenic in humans.

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UNVENTED KEROSENE HEATER EMISSIONS IN MOBILES HOMES: STUDIES ON INDOOR AIR PARTICLES, SEMIVOLATILE ORGANICS, CARBON MONOXIDE, AND MUTAGENICITY

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Human exposure to air pollutants from unvented kerosene heaters in mobile homes was assessed. Eight homes with nonsmokers were monitored, when the kerosene heaters were on and off, for PM_{10}, CO, semivolatile organics, and mutagenicity of particle-phase organics in Salmonella typhimurium. Significant increases in CO and organic concentrations, including polycyclic aromatic hydrocarbons (PAH), nitro PAH, and unburned fuel were found. Four of the eight homes had CO and/or PM_{10} concentrations exceeding the U.S. ambient air standards with the heaters on. Five homes had increased mutagenic activity with the heaters on. These data showed that kerosene heater emissions impact air quality in mobile homes and contain compounds potentially carcinogenic in humans.

INTRODUCTION

Unvented kerosene heaters have been used for space heating in U.S. homes to reduce energy costs. An estimated 15 to 17 million kerosene heaters have been sold in the United States; 33% of these units were sold to mobile home residents (1). Mobile homes usually have small air volume and those built after 1974 have lower ventilation rates than other single-family dwellings (1). The pollutants emitted from unvented kerosene heaters can reach high levels in these mobile homes. Most kerosene heater emission studies reported were conducted in chambers, which are different from the home environment. Previous chamber studies have shown that kerosene heaters are significant sources of inorganic combustion gases, such as NO, NO_2, SO_2, and CO and are sources of fine particles (2). It also has been reported that particle-phase organics are mutagenic in Salmonella typhimurium and that nitro PAH are present in the particle-phase organics (3). Traynor et al. reported that kerosene heaters emit high concentrations of semivolatile organics (4). The objectives of this study were to investigate the effects of kerosene heaters on indoor air quality, and to assess human exposure to air pollutants from unvented kerosene heater emissions in the home environment and relate pollutant levels to the U.S. ambient air standards.

MATERIALS & METHODS

Eight homes located in two trailer parks in Apex, North Carolina were selected for the study. The criteria for home selection were (a) types of kerosene heaters used - four (Homes 1, 2, 3, 6) with convective heaters (21100 kJ/Hr), three (Homes 4,7,8) with radiant heaters (7200-10000 kJ/Hr), and one (Home 5) with a convective/radiant heater (9200 kJ/Hr); (b) heater usage time - homes that used kerosene heaters regularly during the heating season; (c) trailer size - single-wide trailers with a volume of 150-225 m^3; (d) total electric homes - electric cooking and heating systems; and (e) no cigarette smokers living in the home. During the study period, all homes used K1 kerosene fuel containing 0.024% sulfur. Each home was monitored for an average of 6.5 h/day (ranging from 2.6 to 9.5 h/day), 3 days per week for 2 weeks with kerosene heaters on and 2 weeks with the heaters off. Average on-time of the heaters was 4.5 h. The scheduling of on-off status for each home was done in an extended random block fashion, according to a Latin-square study design (5). The participants used kerosene heaters in the manner they normally followed when the heater status was "on" and used only an electric heater, if needed, when the heater status was "off". A Latin-square design analysis was used to test for the effect of the kerosene heater on the pollutant concentrations in homes by comparing average weekly pollutant values after controlling for the effects of week and mobile home. The log values of the average concentrations of pollutants were analyzed by general linear models. All data, with the exception of mutagenicity data, were linearly adjusted for the number of hours per day that the heater was used (4.5 h) and the hours that sampling was conducted (6.5 h).

The pollutants monitored in each home were PM_{10}, semivolatile organics, and CO. The sampling system for PM_{10} and semivolatile organics consisted of a particle sampler equipped with a 10-μm inlet separator and a Teflon-coated glass-fiber filter to collect particles and a XAD-2 canister to collect semivolatile organics at 15 L/min. This sampling system was located in the family room or kitchen at the breathing zone height of a sitting person. The sampler was 1.5-3 m away from the kerosene heater. Sampling usually began as soon as the kerosene heater was turned on and extended 2 h after the heater was turned off to collect the pollutants during decay. For outdoor sampling, high-volume samplers (one at each trailer park) equipped with 10 μm size-selective inlets were sampled simultaneously with the indoor air sampling of homes. Portable SPE CO detectors (General Electric Model 15ECS3C03) were located in the vicinity of the filter/XAD sampling system. During the sampling period the air exchange rate in each home was monitored using the perfluorocarbon tracer technique described previously (6).

The mass of PM_{10} was determined and organic matter was removed from the filter and XAD with dichloromethene. Organic matter from filters collected within each week was pooled for bioassay. Gravimetric (GRAV, for organic compounds with boiling points >300°C) analysis was performed by determining the residual weight of the organics from the XAD samples (7). The XAD samples also were analyzed for total chromatographable organics (TCO) to detect organic compounds with boiling points of 100 to 400°C (7). Quantitative PAH and nitro PAH were determined in the kerosene fuel and the organic extracts from the filter and the XAD samples collected in Home 3 (using a popular convective kerosene heater, Kerosun Omni 105) by GC/MS as reported previously (8). The organic extracts of the emission samples and the fuel samples also were analyzed by GC/MS in the full-scan mode to determine the major components. The particle-bound organics from the filters was tested for mutagenicity in a microsuspension reverse-mutation assay using Salmonella typhimurium TA98 without metabolic activation to detect direct-acting, frameshift mutagens as described previously (9). A sample was considered positive if it showed a dose-response relationship. The slope

values of the linear portion of the dose-response curves were used to obtain the mutagenic activity by the Bernstein linear regression model (10).

RESULTS AND DISCUSSION

The air exchange data in Table I indicate that the air exchange rates were low and similar regardless of the kerosene heater status. Data from eight homes showed no significant effect ($p>0.5$) of kerosene heater use on the indoor PM_{10} concentrations (Table I). Three homes showed higher mean PM_{10} concentrations when the kerosene heaters were on than when they were off. Homes 3 and 8 showed a great increase in PM_{10} concentration with kerosene heaters on and the concentrations exceeded 100 $\mu g/m^3$ (mean concentrations of 122 and 113 $\mu g/m^3$ for Homes 3 and 8, respectively) when the heaters were on. PM_{10} concentrations in these two homes exceeded the 24 h PM_{10} standard (150 $\mu g/m^3$) (11) for at least one day out of the 6 sampling days with the heaters on. The average outdoor PM_{10} concentration of both trailer parks was 18.0 ±2.1 (SEM) $\mu g/m^3$. The data from eight homes showed that the GRAV organic and TCO concentrations of XAD samples were significantly higher ($p<0.01$ and $p<0.025$, respectively) with the heaters on than with the heaters off (Table I). The high semivolatile organic emissions from kerosene heaters in homes were consistent with the findings from our previous chamber study (4).

The effect of kerosene heaters on CO concentrations in the eight homes was highly significant ($p<0.0025$ and $p<0.001$ for CO average concentration during sampling and CO 1-h peak concentration, respectively) as shown in Table I. The CO average concentrations of each week in three homes during the two weeks with the heaters on exceeded the U.S. national ambient air standard for 8-h CO average, 9 ppm (11). Extremely high CO concentrations were found in Home 1 during heater use. In all six sampling days with the heater on, the daily CO average concentrations during sampling in this home exceeded the U.S. national ambient air standard for 8-h CO average and the 1-h CO peak concentrations reached or exceeded the 1-h peak standard of 35 ppm for U.S. ambient air (11) in three of the 6 sampling days. The CO peak concentration in this home reached as high as 51 ppm.

When the samples were tested up to 2 m^3 equivalent of indoor air, dose-response mutagenic activity in the particle-phase organics (filter samples) was shown in 56% of the heater-on days of all eight homes and mutagenic activity was shown in 19% of the heater-off days (Table I). Statistical analysis on the slope values of the dose-response curves showed that the heater effect was significant in a one-tail test ($p<0.025$). Five homes showed an increase in the mean of the mutagenic activity of particle-phase organics with the heaters on, and the mutagenicity of these indoor organics was higher than that of the organic extracts of the outdoor PM_{10} (14.5 ± 2.1 revertants/m^3). Quantitative PAH and nitro PAH analyses of kerosene fuel and organic extracts of the filter and the XAD samples collected from Home 3 with the heater on and off are presented in Table II. These data show that the kerosene fuel contained PAH compounds with four or less aromatic rings. For the emission samples, the XAD sample contained PAH with four or less aromatic rings with the exception of cyclopenta[*c*,*d*]pyrene and the filter sample contained aromatic compounds with up to seven rings. The PAH compounds with four or less rings showed comparable or lower levels in the heater-on days compared to the heater-off days. However, the PAH compounds with four or more aromatic rings were consistently higher with the heater on than with the heater off. These PAH compounds with four or more aromatic rings, including cyclopenta[*c*,*d*] pyrene, benz[*a*]anthracene, benzo[*k*]fluoranthene, benzo[*a*]pyrene, benzo[*g*,*h*,*i*] perylene, and indeno[1,2,3-*c*,*d*]pyrene, have been shown to be animal carcinogens (12). Of these carcinogenic compounds listed above, the XAD sample showed elevated concentrations of cyclopenta[*c*,*d*]pyrene (sevenfold) and benz[*a*]anthracene (threefold) and the filter sample showed

elevated levels of all compounds (some of them by an order of magnitude) when the kerosene heater was on. It has been reported that nitro PAH, including 1-nitronaphthalene and nitrofluoranthenes, are mutagenic in S. typhimurium (13). When the kerosene heater was in use, higher concentrations of 1-nitronaphthalene and nitrofluoranthenes were found in the filter sample. A higher concentration of 1-nitronaphthalene was found in the XAD sample with the heater on. Full-scan GC/MS analysis of organic extracts of the kerosene heater emissions from Home 3 showed that the major compounds present in the filter sample were aliphatic hydrocarbons, fatty acids, and fatty acid esters; and the major compounds present in the XAD samples were aliphatic hydrocarbons, pienes, alkylbenzenes, chlorobenzenes, and alkylnaphthalenes. Most of the compounds found in the XAD sample also were found in the kerosene fuel. These results suggested that some of the organic components present in the XAD samples were unburned, evaporated kerosene fuel.

The number of kerosene heaters investigated in this study was limited. It is difficult to draw a conclusion about the effect of heater type on production of pollutants in homes. We observed that the convective heater Kerosun Omni 105 (used in Home 3) emitted high PM_{10} that showed high mutagenic activity. Home 6 also used the same model of heater but showed low emission of PM_{10}. One possible reason for the low emission may be that the heater in Home 6 was located in front of the air intake of the central heating system and the particles were filtered prior to entering the living area where the sampling took place. The radiant heater, Happy Radiant II, used in Home 8 also emitted high levels of PM_{10} that showed high mutagenic activity. The heater that showed high concentrations of CO was a convective heater, the Tokyokuni Dual Combustion model.

CONCLUSION & RECOMMENDATIONS

We found increased CO concentrations and organic emissions that contained PAH and nitro PAH in mobile homes where kerosene heaters were used. High concentrations of TCO and GRAV organics were emitted from kerosene heaters in the homes. Chemical analysis suggested the presence of unburned, evaporated kerosene fuel in the semivolatile organic emissions. Four of eight homes had CO and/or PM_{10} concentrations that exceeded the U.S. national ambient air standards when the kerosene heaters were on. Five homes had increased mutagenic activity when the kerosene heaters were in use. This study indicated that kerosene heater emissions can significantly impact indoor air quality, and the organic emissions from the kerosene heater contained compounds potentially carcinogenic in humans.

Further investigations are needed, including determination of the effect of the type of kerosene heater on indoor air pollution, evaluation of heater maintenance on reducing pollutant levels and the effect of ventilation, such as opening windows, on reducing pollutant levels. This information will be useful in providing guidance to consumers on heater selection and maintenance. In addition, studies are needed on (1) health effects of inhaling high concentration of semivolatile organics, including PAH and the unburned kerosene fuel, (2) to investigate kerosene heater emissions in mobile homes when residents operate the heaters for a longer period of time as a primary heating source, and (3) to investigate the synergistic effects of pollutants from kerosene heaters and other indoor sources. [The research described in this paper has been reviewed by the Health Effects Research Laboratory, U.S. EPA and approved for publication. Approval does not signify that the contents necessarily reflect the views and policies of the Agency nor does mention of trade names constitute endorsement for use].

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Table I. Summary of Indoor Pollutant Concentrations and Air Exchange Rates in Eight Mobile Homes with Kerosene Heaters On and Off during the Four-Week Sampling Period

	Mean ± SEM			
	Heater On		Heater Off	
	N[a]	Values	N	Values
PM10 ($\mu g/m^3$)	45	73.7 ± 7.3	46	56.1 ± 5.7
XAD•GRAV (mg/m^3)	45	0.96 ± 0.15	46	0.46 ± 0.07
XAD•TCO (mg/m^3)	45	3.31 ± 0.45	46	2.12 ± 0.22
CO average (ppm)	30	7.4 ± 1.4	37	1.4 ± 0.3[b]
CO peak (ppm)	30	11.5 ± 2.5	37	1.5 ± 0.3[b]
Mutagenicity: % Positive	16	56[c]	16	19[c]
Revertants/m^3 (of the positive)	9	23.3 ± 4.8	3	14.1 ± 1.9
Air exchange rate (Air exchange/h)	43	0.47 ± 0.05	46	0.48 ± 0.05

[a] N = Number of sampling days or number of samples analyzed.

[b] Data from seven homes, not including data from Home 7.

[c] Samples were tested up to 2 m^3 of air.

Table II. PAH and Nitro PAH Analysis of K1 Kerosene and Indoor Air Samples from Home 3 with a Kerosene Heater On and Off

Compounds	Particle-Phase Organics (filter samples) ng/m^3		Semivolatile Organics (XAD Samples) ng/m^3		Kerosene Fuel ng/μl
	Off[a]	On[b]	Off	On	
Naphthalene	0.47	0.24	2300	950	940
Acenaphthylene	1.9	0.021	19	68	8.6
Phenanthrene	4.9	0.82	43	33	64
Fluoranthene	1.6	1.1	14	9.5	9.9
Pyrene	1.4	0.76	8.3	12	25
Cyclopenta[c,d]pyrene[c]	0.11	1.5	0.38	2.7	BDL[d]
Benz[a]anthracene[c]	0.17	1.3	0.55	1.5	BDL
Chrysene	0.39	1.3	1.1	1.8	BDL
Benzofluoranthenes[c]	0.45	5.5	BDL	BDL	BDL
Benzo[a]pyrene[c]	0.24	2.0	BDL	BDL	BDL
Indeno[1,2,3-c,d]pyrene[c]	0.15	1.3	BDL	BDL	BDL
Benzo[g,h,i]perylene[c]	0.22	3.7	BDL	BDL	BDL
Coronene	0.069	2.3	BDL	BDL	BDL
1-Nitronaphthalene	0.008	0.010	0.24	1.5	NA[e]
2/3-Nitrofluoranthene	0.068	0.10	BDL	BDL	NA
1-Nitropyrene[c]	0.081	0.057	BDL	BDL	NA
Dinitropyrenes[c]	BDL	BDL	BDL	BDL	NA

[a] Data from the samples collected in the first week of the four-week sampling period.
[b] Data from the samples collected in the fifth week following the four-week sampling period provided additional heater-on days for detailed chemical analysis.
[c] Compounds showing evidence of animal carcinogenicity (12, 14).
[d] BDL = Below detection limit.
[e] NA = Not analyzed.

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Key words: kerosene heater emissions, PM_{10}, organic emissions, carbon monoxide, total chromatographable organics, gravimetric organics, polycyclic aromatic hydrocarbons (PAH), nitro PAH, mutagenicity, Salmonella typhimurium.

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